

# ALUMINUM COLLOID FORMATION BY HOMOGENEOUS NUCLEATION IN A SUPERSONIC NOZZLE

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## INTRODUCTION

The use of the spent booster rocket tankage as a propellant for electric spacecraft has been shown to be very desirable<sup>7.1</sup>. For many missions, the payload could be more than doubled if this spent tankage could be used. Research has been directed toward development of a charged-particle electrostatic thruster that can operate with aluminum propellant. This section contains the principles involved in aluminum colloid formation in a supersonic nozzle. The section also includes parameter studies made on the important variables to help determine the optimum nozzle design.

## THEORY AND METHOD OF ANALYSIS

### Condition for Condensation to Form During Supersonic Expansion

The rate of change of pressure with respect to temperature during an isentropic expansion is given by:

$$\frac{dp}{dT} = \frac{C_p}{R} \frac{P}{T} \quad (7.1)$$

where  $R$  is the gas constant of the vapor, and  $C_p$  is the specific heat of the vapor.

The rate of change of the saturation pressure with temperature is given by the Clausius - Clapeyron equation:

$$\frac{dP_{\infty}}{dt} = \frac{h_{fg}}{(V_v - V_L)T} \quad (7.2)$$

where  $V_L$  and  $V_v$  is the specific volume of the liquid and the vapor phases and  $h_{fg}$  is the latent heat of vaporization since  $V_v - V_L \doteq V_v$  and  $V_v = 1/\rho_v$  where  $\rho_v$  is the density of the vapor. Using the perfect gas law,  $\frac{P}{RT} = \frac{1}{V_v}$ , the Clausius - Clapeyron equation can be rewritten as:

$$\frac{dP_{\infty}}{dT} = \frac{P}{T^2} \frac{h_{fg}}{R} \quad (7.3)$$

Comparing this to the rate of change of pressure with temperature with isentropic expansion:

$$\frac{dP_{\infty}/dT}{dP/dT} = \frac{Ph_{fg}/T^2R}{C_p P/RT} = \frac{h_{fg}/T}{C_p} \quad (7.4)$$

The ratio for aluminum (at boiling point) is:

$$\frac{dP_{\infty}/dT}{dP/dT} = \frac{70,200 \text{ g-cal/g-atom}}{(2450 + 273)^{\circ}\text{K}/(5.1 \text{ g-cal/g-atom}^{\circ}\text{K})} = 5.04$$

Thus, when  $h_{fg}/T > C_p$ , the saturation vapor pressure decreases with decreasing temperature more rapidly than does the static pressure in an isentropic expansion.

Since

$$\frac{h_{fg}}{T} = 5.04 C_p$$

aluminum should condense during a supersonic expansion.

The supersaturation ratio,  $P/P_{\infty}$ , continually increases until saturation, and eventually supersaturation is obtained. This is shown on Figure 7.1 below.

If  $h_{fg}/T < C_p$ , an isentropic compression would be necessary for approach to saturation condition.

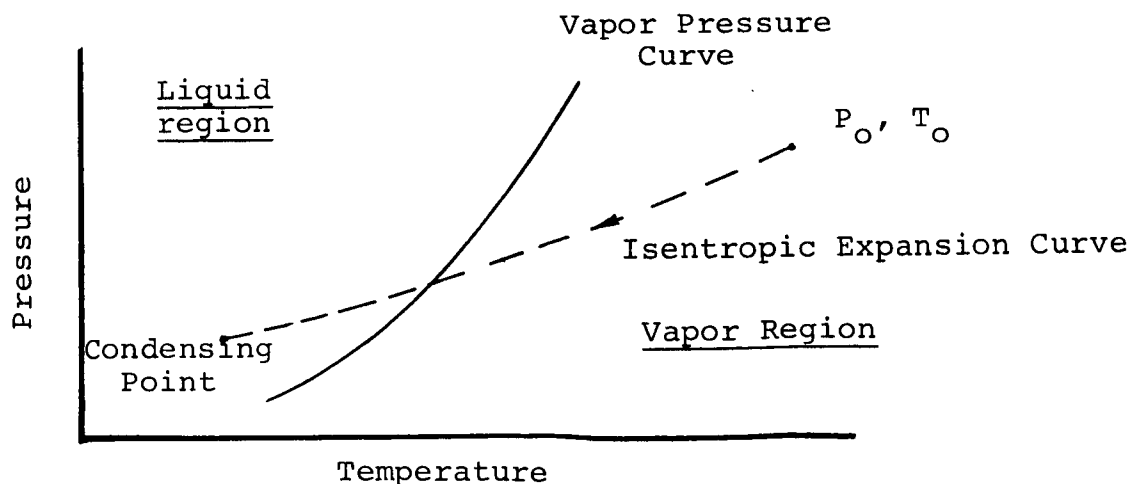


Figure 7.1. The Comparison of Vapor Pressure Curve with an Isentropic Expansion for a Condensing Vapor

## Formation of the Critical Size Droplet (or Colloid)

The energy required for droplet formation is given by the Gibbs free energy:

$$\Delta G = Vdp - s dT$$

$V$  = specific volume,  $s$  = entropy

The minimum change of free energy for vapor molecules in reducing their pressure  $P$  to the saturation value  $P_\infty$  is:

$$\Delta G_1^* = 4/3\pi r^3 \rho_L RT \ln P/P_\infty$$

The surface free energy for creation of small drops is:

$$\Delta G_2^* = 4\pi (r)^2 \sigma$$

Net change in free energy is:

$$\Delta G = \Delta G_2 - \Delta G_1$$

or

$$\Delta G = 4\pi (r^2) \sigma - 4/3\pi r^3 \rho_L RT \ln (P/P_\infty)$$

The minimum size (critical radius,  $r^*$ ) droplets that can exist is given when  $\frac{\Delta G}{dr} = 0$  which can be thought of as the maximum activation energy required for the formation of a droplet. This is shown on the figure below:

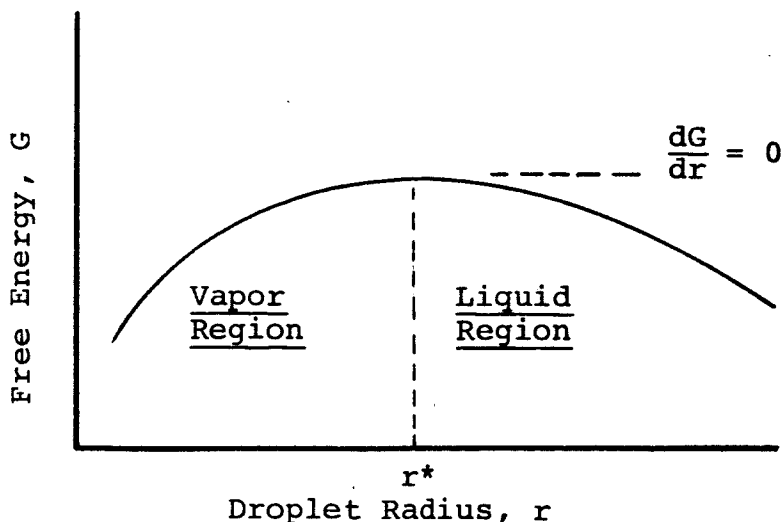


Figure 7.2. Free Energy Versus Droplet Radius

Taking the derivative of the equation above:

$$8\pi r^* \sigma = 4\pi r^2 \rho_L RT \ln (P/P_\infty)$$

or

$$r^* = \frac{2\sigma}{\rho_L RT \ln (P/P_\infty)} \quad (7.5)$$

Note that the unknown in the equation is the ratio of  $P/P_\infty$  where the critical size is found. This ratio can be determined by test.

Example: The critical droplet size for aluminum condensation versus stagnation pressure,  $P$ , and temperature  $T$ , was computed using the following data:

surface tension	= 520 dyne/cm
density of liquid	= 2.26 g/cm <sup>3</sup>
gas constant, $R$ ,	= $8.317 \times 10^7$ ergs/ <sup>o</sup> K/mole
atomic weight	= 26.97 g/mole

$$r^* = \frac{2(520)}{(2.26) \frac{(8.317 \times 10^7)}{26.97} T \ln (P/P_\infty)}$$

$$r^* = \frac{1.49 \times 10^{-4}}{T \ln (P/P_\infty)}$$

The value of  $r^*$  is shown for various static temperatures and  $P/P_\infty$  ratios on Figure 7.3.

#### Rate of Droplet Formation

A distribution of embryos exists which satisfies a Boltzman type of distribution relation:

$$n_g = N_a e^{-\Delta G/KT} \quad (7.6)$$

where  $n_g$  is the number of cluster of  $g$  molecules, and  $K$  is Boltzman constant.

Similarly, the rate of droplet formation,  $J$ , can be written:

$$J = \left(\frac{P}{KT}\right)^2 \frac{1}{\rho_L} \sqrt{\frac{2\sigma M}{\pi N_a}} e^{-4\pi r^{*2}\sigma/3KT} \quad (7.7)$$

where  $M$  = Molecular weight,  $N_a$  = Avagadro's number,  $P/KT\rho_L$  is proportional to number of molecules,  $(2\sigma M/\pi N_a)^{1/2}$  is proportional to the diffusion rate. Note the strong dependence of nucleation rate on surface tension  $\sigma$ , especially since  $r^*$  is also proportional to  $\sigma$ .

#### PARAMETER STUDY FOR DESIGN OF A PROTOTYPE OF THE SUPERSONIC ALUMINUM COLLOID NOZZLE

##### Study of Plenum Conditions Required

This study was conducted to determine how the choice of stagnation conditions affects the size and initial ratio of particle nucleation. For this initial study, it was assumed that the expansion through the nozzle was isentropic and that the vapor behaved as a perfect gas. This assumption is good until a substantial fraction of the aluminum condenses.

The ratios of the stagnation pressure to the static pressure, the stagnation temperature  $T_0$  to the static temperature are a function of the Mach number,  $M$ , by the following relationships:

$$\frac{P}{P_0} = \left(1 + \frac{\gamma-1}{2} M^2\right)^{\gamma/(\gamma-1)} \quad (7.8)$$

$$\frac{T_0}{T} = 1 + \frac{\gamma-1}{2} M^2 \quad (7.9)$$

where  $\gamma$  = specific heat ratio, where the relationship between  $P$  and  $T$  can be written as:

$$P = P_0 \left(\frac{T}{T_0}\right)^{\gamma/(\gamma-1)} \quad (7.10)$$

Thus, as the vapor is expanded through the nozzle, the pressure  $P$ , and temperature,  $T$ , decrease. The vapor pressure,  $P_\infty$ , for aluminum was found to decrease faster than the static pressure

during an isentropic expansion. When the ratio of  $P/P_\infty$  at a given temperature is large enough to overcome the energy barrier between vapor and liquid phase (see Figure 7.2), droplets of critical radius should form. The critical ratio of  $P/P_\infty$  is believed to be about 6 for aluminum. Since the static temperature,  $T$ , and pressure,  $P$ , are only a function of the stagnation conditions,  $T_0$ ,  $P_0$ , and the Mach number,  $M$ , (as shown by Equations (7.8) and (7.9), the choice of stagnation conditions will also determine the velocity at which condensation will occur. The effect of the choice of the plenum stagnation pressure and temperature on the condensate was studied using a design considered in previous work<sup>7.1</sup>. The design consisted of 2.6 mm throat diameter and plenum temperature of 1800°K and pressure of 0.005 atmosphere (3.8 mmHg). The flow-rate,  $\dot{m}$ , through the nozzle can be written as:

$$\dot{m} = \frac{A^* P_0}{\sqrt{T_0}} \sqrt{\frac{\gamma}{R}} \left( \frac{2}{\gamma + 1} \right)^{\frac{\gamma + 1}{2(\gamma - 1)}} \quad (7.11)$$

From Equation (7.11), it is noted that, for a fixed nozzle throat area  $A^*$ , as long as the ratio  $P_0/\sqrt{T_0}$  is held constant, the flow rate,  $\dot{m}$ , will be unchanged. To study the velocity of which condensation would form, various values of  $T_0$  were chosen and  $P_0$  was computed to hold the ratio of  $P_0/\sqrt{T_0}$  constant. The Mach number,  $M$ , where condensation would begin was then computed as a function of the saturation ratio  $P/P_\infty$ . The results of this analysis are shown on Figure 7.4. The importance of the stagnation temperature on the velocity is clearly shown on Figure 7.4. A stagnation pressure of 4 mmHg (.0053 atmospheres) pressure, a stagnation temperature of at least 2000°K is shown to be required to get the condensate to sonic velocity.

The rate of droplet formation,  $J$ , corresponding to the above nozzle design, was computed using Equation (7.7) and it shows  $J$  as a function of stagnation pressure, stagnation temperature, and saturation pressure ratio on Figure 7.5. The rate of droplet formation is shown on Figure 7.5 and was found to be too low for

a nozzle design suitable for test in the CSU vacuum chamber. This rate of formation would require too large a nozzle to get a practical condensation fraction.

#### Study of Effects of Pressure and Temperature on Condensation Rates and Droplet Size

A general parameter study was made to determine the effects of the temperature and pressure on the condensation formation rates and the critical droplet sizes. A wide range of pressure and temperatures was chosen and an analysis was made using Equations (7.6) and (7.7). To have a high rate of droplet formation (see Figure 7.6), it was found that the static pressure where the condensate forms must be sufficiently large. It also should be noted from Figure 7.6 that the droplet sizes increase very rapidly (for a given static temperature) while the condensation rate falls rapidly because of inadequate static pressure.

#### Study of Nozzle Length Required

One of the basic requirements for the colloid nozzle for use in space is keeping the size as small as possible. The nozzle however, must be large enough for the heat of condensation to be removed or else the static temperature will increase quickly and the rate of condensate will decrease as shown on Figure 7.6. (More will be said on removal of the condensation heat later.) The length of the nozzle required for the aluminum vapor to condense,  $L$ , can be estimated as follows:

$$L = v_c t_c \quad (7.12)$$

where  $v_c$  is the velocity of the condensate and  $t_c$  is the time for the aluminum to condense. This time for condensation to form can be approximated (assuming the rate of formation,  $J$ , and the critical droplet size,  $r^*$  remains constant through the nozzle. Thus  $t_c$  can be written as:

$$t_c = \frac{\rho_v}{J \frac{4}{3}\pi r^{*3} \rho_L} \quad (7.13)$$

where  $\rho_v$  = the density of the aluminum vapor and  $\rho_L$  = the density of the aluminum liquid. The velocity of the aluminum vapor,  $v$ , can be written as:

$$v = M \sqrt{\gamma RT} \quad (7.14)$$

Where  $M$  is the Mach number where the condensate forms, and  $\sqrt{\gamma RT}$  is the velocity of sound in the aluminum vapor. If the temperature at which condensate forms is  $1000^\circ\text{C}$ , the velocity of sound in the aluminum vapor would be 805 m/sec. Assuming that the condensate forms at  $M = 1.25$ , the velocity of the condensate would be approximately 1000 m/sec. An analysis was made to roughly determine the length of the nozzle required for complete condensation of the aluminum vapor. This analysis assumed that:

1. The condensation rate  $J$  (shown on Figure 7.6) remained constant.
2. The velocity of the condensation was constant and equal to 1000 m/sec.

The distances required for condensation as a function of the static temperature and static pressure are shown on Figure 7.7. For testing in the CSU vacuum tank, the length would probably be between 10 to 100 cm. Using Figure 7.7, the pressure and the temperature required for an aluminum colloid nozzle test can be determined.

#### CONDENSATION HEAT REMOVAL

When the vapor in the nozzle forms the condensate, the latent heat of vaporization is released. The latent heat for liquid metals is very great compared to the specific heat. For example, the specific heat of liquid aluminum is  $6.7 \frac{\text{g cal}}{\text{g atom}^\circ\text{C}}$  while the latent heat of vaporization is  $70,200 \frac{\text{g cal}}{\text{g atom}}$ . Thus, if the heat were not removed, the temperature of the aluminum droplet would greatly increase and the droplet would quickly re-evaporate. Furthermore, if the temperature of the aluminum vapor increases, the rate of condensation decreases rapidly (see Figure 7.6). It would be



highly desirable, not only to remove the latent heat of the condensate, but also to lower the static temperature of the uncondensed aluminum vapor to help increase the rate of droplet formation. Two methods of removing the latent heat from the particles are being studied at present. These methods are cooling by the vapor expansion and cooling by gas injection.

### Cooling by Vapor Expansion

It is possible to cool the vapor by expansion in the nozzle and to use this cooled vapor to remove the heat of the condensate droplets. The basic equation for this expansion cooling process is:

$$T_o = T + \frac{v^2}{2C_p} \quad (7.15)$$

Thus from Equation (7.15) it is seen that, in order to lower the static temperature,  $T$ , an increase of velocity,  $v$ , is required.

When the condensate starts to form in the nozzle the stagnation temperature of the stream is increased, as given by the equation below:

$$\dot{m}C_p (T_{02} - T_{01}) = \dot{m}_L h_{fg} \quad (7.16)$$

where  $T_{01}$  and  $T_{02}$  are the respective stagnation temperatures upstream and downstream of the condensate,  $\dot{m}$  is the total mass flow,  $\dot{m}_L$  is the mass flow rate of the condensing liquid, and  $h_{fg}$  is the latent heat per unit mass of the condensing liquid. Equation (7.16) may be rearranged to give:

$$\frac{T_{02}}{T_{01}} = 1 + \frac{\dot{m}_L}{\dot{m}} \frac{h_{fg}}{C_p T_{01}} \quad (7.17)$$

The nozzle area charge required for the vapor expansion cooling of the condensate was studied for expansion having three basic criteria which are:

1. Constant Mach number of the vapor.
2. Constant velocity of the vapor.
3. Constant static temperature.

Using the criteria above, analyses were made to determine the area changes required to remove the condensation heat from various stagnation temperature ratios. The equations used in the analyses were derived from differential equations given in Chapter 8 of Reference 7.2. The equations are stated below.

The area charge required for condensation heat removal with a constant vapor Mach number is given by:

$$A_2/A_1 = \left( \frac{T_{02}}{T_{01}} \right)^{\frac{1 + \gamma M^2}{2}} \quad (7.18)$$

The area charge required for condensation heat removal with a constant vapor velocity is given by:

$$\frac{A_2}{A_1} = \left( \frac{T_{02}}{T_{01}} \right)^{1 + \frac{\gamma - 1}{2} \bar{M}^2} \quad (7.19)$$

where  $\bar{M}$  is the average Mach number between point 1 and 2.

The area charge required for condensation heat removal with a constant vapor static temperature is given by:

$$\frac{A_2}{A_1} = \left( \frac{T_{02}}{T_{01}} \right)^{\frac{(\gamma \bar{M}^2 - 1) (1 + \frac{\gamma - 1}{2} \bar{M}^2)}{(\gamma - 1) \bar{M}^2}} \quad (7.20)$$

A parameter study was made using Equations (7.18), (7.19), and (7.20) and the results of these analyses are shown on Figures 7.8, 7.9 and 7.10 respectively.

It should be noted that Equations (7.18), (7.19) and (7.20) assume that the vapor acts as a perfect gas and that the fraction of the condensing gas is small. When the condensation is a large fraction of the vapor, an additional term must be added to the above equation. More detailed analyses of the condensate vapor expansion cooling would require a computer program which is under study at present. The answer given by the above method, however, is satisfactory for this preliminary study.

To show the order of magnitude of the area charge required to cool the nozzle during a condensation process, a typical numerical example is given below.

Example Calculation of Cooling Condensation by Vapor Expansion.

This calculation is to determine the area charge of the velocity and temperature of the condensate cooled by vapor expansion. Given aluminum vapor condensation has started at a static pressure of .03 atmospheres and static temperature of  $1200^{\circ}\text{C}$ , ( $1473^{\circ}\text{K}$ ) and a Mach number of 1.25.

To find:

- a. The stagnation pressure and temperature conditions.
- b. The rate of droplet formation and droplet size.
- c. The velocity of colloidal particle at condensation start.
- d. The length required for 20% of the vapor to condense.
- e. With 20% of the vapor condensing, find the area change, static temperature, and velocity of vapor for the following conditions:
  1. Constant Mach number during condensation.
  2. Constant velocity during condensation.
  3. Constant static temperature during condensation.

Solution:

- a. The stagnation pressure and temperature can be found using Equations (7.8) and (7.9):

$$T_{01}/T_1 = 1 + \frac{\gamma - 1}{2} M^2 = 1 + \frac{1.67 - 1}{2} (1.25)^2 = 1.523$$

$$T_{01} = (1473) (1.523) = 2250^{\circ}\text{K}$$

$$P_{01}/P_1 = 1 + \frac{\gamma - 1}{2} M^2 \gamma / (\gamma - 1) = (1.523)^{1.67/(1.67 - 1)} = 2.85$$

$$P_{01} = (.03) (2.85) = .0855 \text{ atmospheres}$$

- b. The rate of droplet formation and droplet can be found using Figure 7.6 which gives:

$$J = 1.6 \times 10^{21} \text{ particles/cm}^3 \text{ sec}$$

$$r^* = 3.5 \text{ \AA}^0$$

c. The velocity at the start of condensation can be computed using Equation (7.14):

$$v_1 = M \sqrt{\gamma RT} = 1.25 \left[ 1.67 \frac{(8320)}{26.97} (1473) \right]^{\frac{1}{2}}$$

$$v_1 = 1.25 (870) = 1090 \text{ m/sec}$$

d. The fraction of aluminum vapor condensing per cm length can be estimated as follows:

The density of the aluminum vapor,  $\rho_v$  at start of condensation is equal to:

$$\rho_v = \frac{P}{RT} = \frac{(.03)(26.97)}{(82.05)(1473)} = .67 \times 10^{-5} \text{ g/cm}^3$$

The mass flow rate of the condensate,  $\dot{m}_L$  is equal to:

$$\dot{m}_L = J(4/3\pi r^3) \rho_L = (1.6 \times 10^{21}) \left[ (4/3)\pi (3.5 \times 10^{-8})^3 \right] 2.26$$

$$\dot{m}_L = .65 \text{ g/cm}^3 \text{ sec}$$

The distance required for 20% of the aluminum vapor to condense  $L_{.20}$  (assuming constant velocity and constant condensation rate) is given by Equation (7.12) as follows:

$$L_{.20} = \frac{(.20)(\rho_v)v_1}{\dot{m}_L} = \frac{.20(.67 \times 10^{-5})(1.09 \times 10^5)}{.65} = .225 \text{ cm}$$

e. The stagnation temperature rise,  $\Delta T$ , due to condensation fraction,  $\dot{m}_L/\dot{m}$  is equal to:

$$\Delta T = \frac{\frac{\dot{m}_L}{\dot{m}} h_{fg}}{C_{p_v}} \quad (7.16)$$

The  $\Delta T$  for the aluminum vapor with the 20% condensing is:

$$\Delta T = \frac{(.20)(70,200)}{5.1} = 2760^\circ \text{K}$$

Thus:  $T_{02} = T_{01} + \Delta T$

$$T_{02} = 2250 + 2760 = 5010^{\circ}\text{K}$$

#### Case 1. Constant Mach number

For constant Mach number, the static temperature at the end of 20% condensation,  $T_2$ , can be solved for as follows:

$$\frac{T_{02}}{T_{01}} = \frac{T_2}{T_1} \quad (7.21)*$$

$$\frac{5010}{2250} = \frac{T_2}{1473}$$

$$T_2 = (2.223)(1473) = 3280^{\circ}\text{K}$$

The velocity at the end of 20% condensation,  $v_2$ , is found using Equation (7.14):

$$v_2 = 1.25 \left[ 1.67 \frac{(8320)}{26.97} (3280) \right]^{\frac{1}{2}} = 1,625 \text{ m/sec}$$

The area change required to maintain a constant Mach number during heating is given by Figure 7.8. Reading this figure at  $T_{02}/T_{01} = 2.223$  and Mach number of 1.25, the area ratio required is:

$$A_2/A_1 = 4.2$$

#### Case 2. Constant velocity

The static temperature after 20% condensation is solved for using Equation (7.15) in the following form:

$$\frac{T_{02}}{T_{01}} = \frac{T_2 + \frac{v_2^2}{2C_p}}{T_1 + \frac{v_1^2}{2C_p}} \quad (7.22)$$

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\* See Equation (7.11) of Reference 7.2.

Since  $T_{02}/T_{01} = 2.223$ , and  $V_1 = V_2 = 1090$  m/sec,  $T_2$  can be solved for as follows:

$$\frac{V_1^2}{2C_p} = \frac{(1090 \text{ m/sec})^2}{2(5.1 \frac{\text{g} \cdot \text{cal}}{\text{g atom}^\circ\text{K}} (4.184 \frac{\text{joules}}{\text{cal}}) (1000 \text{ g/kg})} = 753^\circ\text{K}$$

substituting into Equation (7.22) gives:

$$2.223 = \frac{T_2 + 753}{1473 + 753}$$

and

$$T_2 = 4117^\circ\text{K}$$

The area change for this constant velocity expansion is given on Figure 7.9. However, before the curve can be used, the average Mach number  $\bar{M}$  between points 1 and 2 must be solved. At point 2, the Mach number can be solved for, using Equation (7.14):

$$1090 = M_2 \left[ 1.67 \left( \frac{8320}{26.97} \right) (4117) \right]^{\frac{1}{2}}$$

$$M_2 = 1.335$$

The average Mach number  $\bar{M}$  is approximately equal to:

$$\bar{M} = \frac{M_1 + M_2}{2} = \frac{1.25 + 1.335}{2} = 1.292$$

Thus, using Figure 7.19, the area change ratio is obtained:

$$A_2/A_1 = 3.48$$

### Case 3. Constant static temperature

For constant static temperature during condensation, the velocity  $V_2$  and the area ratio  $A_2/A_1$  are computed below. To solve for  $V_2$ , Equation (7.22) can be written as follows:

$$2.223 = \frac{1473 + \frac{V_2^2}{(2)(5.1) \frac{26.97}{(4.184)(1000)}}}{1473 + 753}$$

Therefore:

$$V_2 = 2350 \text{ m/sec}$$

The Mach number at point 2 is equal to:

$$M_2 = \frac{2350}{870} = 2.70$$

and

$$\bar{M} = \frac{1.25 + 2.70}{2} = 1.975$$

Using Figure 7.10, the required area ratio is obtained:

$$A_2/A_1 = 49.5$$

The results of this example and discussion comparing the three cases of expansion cooling during condensation heating are summarized below:

	Velocity at Point 2 m/sec	Static Temperature at Point 2 °K	Area Ratio Required $A_2/A_1$
Constant Mach No.	1625	3280	4.2
Constant Velocity	1090	4117	3.48
Constant Static Temperature	2350	1473	49.5

It is noted that, due to the high static temperature rise for the constant Mach number and the constant velocity cases, the condensate would have evaporated; therefore, there would not have been 20% condensation as assumed. The actual amount of condensate could be solved for by trial and error using the similar method as above.

The area ratio of the constant static temperature case is so great that a shock would probably be formed in the nozzle since the length of the condensation region is so short. For an optimum design of the nozzle, the condensation rate must be balanced with

the length required for cooling of the condensate. It can therefore be concluded that the constant static temperature expansion cooling of the condensate would give the highest amount of condensate but would require a longer nozzle.

It should also be noted that the vapor-expansion cooling of the condensate at constant static temperature yields the highest exit velocity. This gain of velocity helps offset the requirement for a longer nozzle.

### Cooling by Gas Injection

The injection of a non-condensing cooling gas in the condensing vapor region has been considered as a method of cooling. The gas would be injected into the nozzle at the point where the condensation is desired. The analysis of the nozzle upstream of the injection point would be done in a manner similar to the one above. The injected gas could be cooled very much below the temperature of the nozzle condensing gas and could be injected at high velocity. When the injected gas meets the condensing vapor, a very large fraction (perhaps all) of the condensate heat could be removed. This would mean that it could be possible to have all the gas condensing at once.

The choice of the injection gas is under study, but at present, helium and hydrogen seem the most promising because of the following reasons:

1. They have a high specific heat, which would release the amount of gas required.
2. They have a high ionization potential, helium 24.6 ev, hydrogen 13.6, compared to aluminum ionization energy of 5.98 ev. This means that they would not interfere with the charging of the aluminum colloidal particles if the ionization energy were kept below 13.6 ev (24.0 ev).
3. They have low molecular weight which means a high thermal acceleration is possible. Therefore, this injected gas would help increase the velocity of the droplets.

The decision between helium and hydrogen would depend on whether hydrogen would be reactive with the aluminum vapor or the walls of the nozzle. If hydrogen were not to react, it would probably be the best choice since it has higher specific heat and



and lower molecular weight than helium. Also hydrogen would be easier to store since its freezing point is higher.

There are several advantages and disadvantages of the gas injection system of cooling the condensate as compared with the vapor expansion systems. Table 7.1 compares the two methods of cooling the condensate.

There also is the possibility of using both the vapor expansion and the gas injection systems on the same nozzle. This would be done by first cooling the gas by vapor expansion (to save on the injection gas) and then using the gas injection systems. This type of analysis is being considered in the development of a computer code.

A computer code is being developed to handle analysis of the nozzle from the plenum through the condensation and colloidal particle growth. This code will give the nozzle contour required for a given exit velocity and condensate fraction. The code will be able to determine the colloidal particle size along the nozzle and its velocity. (More will be said about the particle velocity below.) Provision will be made in the code for gas injection at any point along the nozzle.

The colloidal particles would grow, due to vapor condensating on them and to collisions between the particles. As the particles grow, the velocity difference between the vapor molecules and the colloidal particles will increase. The method of analysis which will be used in the code is developed below:

## RELATIVE VELOCITY BETWEEN THE CONDENSATE AND THE GAS

### Theory

The drag force,  $F_D$ , acting on a spherical body with a velocity relative to a gas may be represented by:

$$F_D = C_D A \frac{1}{2} \rho_V (\Delta v)^2 \quad (7.23)$$

$F$  = force acting on particle

$C_D$  = drag coefficient

$\rho_V$  = gas density

$A$  = frontal area of condensate =  $\pi r^2$

$\Delta v$  = relative velocity between gas and particle =  $v_g - v_c$

$\mu_g$  = viscosity of the gas

The acceleration of the particle is determined by the force acting on its mass:

$$F_a = ma = 4/3\pi\rho_s r^3 a = 4/3\pi\rho_s r^3 \frac{dv_c}{dt} \quad (7.24)$$

$\rho_s$  = density of particle,  $v_s$  its velocity

$r$  = radius of particle

$a$  = acceleration of particle =  $dv_c/dt$

Equation (7.23) and (7.24)

$$4/3\pi\rho_s r^3 \frac{dv_s}{dt} = C_D A (1/2 \rho_V (v_g - v_s)^2)$$

or

$$\frac{dv_s}{dt} = \frac{3C_D \rho_V}{8\rho_s r} (v_g - v_c)^2 \quad (7.25)$$

Since  $C_D$  is a function of  $(v_g - v_c)$ , its equation must be substituted into Equation (7.25) before it can be integrated.

Example: for small velocity lags  $R_e < 2$ , Stokes Law can be used where:

$$C_D = 24/R_e \quad (7.26)$$

Where the Reynolds Number,  $R_e$  is:

$$R_e = \frac{2(v_g - v_c) r}{\mu_g} \quad (7.27)$$

substituting Equation (7.27) into (7.26):

$$C_D = \frac{12\mu_g}{(v_g - v_c) r} \quad (7.28)$$

Substituting Equation (7.28) into (7.25):

$$\frac{dv_c}{dt} = \frac{9\mu_g}{2r^2} \frac{(v_g - v_c)}{\rho_s} \quad (7.29)$$

Similarly, the drag coefficient can be written as a function for all Reynolds numbers. For example: if  $2 < R_e < 5$ , use Oseen's equation:

$$C_D = \frac{24}{R_e} + (1 + 3/16 R_e)$$

#### Method of Iteration to be Used in the Code

If condensation starts at point 1, then at this point it is assumed  $v_s = v_g$ .

The radius,  $r$ , will change with time as the particle size increases. At point 1, the particle will have its critical radius,  $r^*$ . It is also known that:

$$\frac{dv_c}{dt} \leq \frac{dv_g}{dt} \quad (7.30)$$

Thus, at point 1, assume  $\frac{dv_c}{dt} = \frac{dv_g}{dt}$  :

Thus, since  $\frac{dv_g}{dt}$  is known from the nozzle design,  $(v_g - v_c)$  can be solved using either Equation (7.25) or (7.29) depending on the Reynold number. Using the value for  $(v_g - v_c)$ , the acceleration force on particle,  $F_a$  can be found. Compare this to the drag force on the particle,  $F_D$ . If  $F_a > F_D$ , reduce estimate of  $\frac{dv_c}{dt}$  until  $F_D = F_a$  using method below:

$$\left(\frac{dv_c}{dt}\right)_{\text{new}} = \left(\frac{dv_c}{dt}\right)_{\text{old}} \left(\frac{F_D}{F_a}\right)_{\text{old}}^\alpha \quad (7.31)$$

Note that the value of  $\alpha$  will be determined by the value which makes the code converge the fastest. This value will also depend on the rate of expansion of the nozzle. It is estimated that,  $\alpha$  will have its best value between 0.25-0.50.

#### CONCLUSION AND FUTURE WORK

This report shows the fundamental principles involved in aluminum colloidal formation by homogeneous nucleation in a supersonic nozzle. The design theory and requirements are

discussed from the plenum, where the solid aluminum would be vaporized, through the region of condensation. The following topics were discussed:

1. The theory of homogeneous nucleation
  - a. Critical size of the aluminum colloidal particles.
  - b. Rate of colloid formation of the aluminum colloidal particles and velocity of these particles.
  - c. Length of the nozzle required for condensation.
2. Condensate heat removal
  - a. The cooling of the condensate by the vapor expansion.
  - b. The cooling of the condensate by gas injection.
3. The analysis of the velocity of the condensate compared to the vapor velocity.

The preliminary analysis of the design of the supersonic nozzle for aluminum colloid formation by homogeneous nucleation yielded the following conclusion.

The choice of the plenum temperature and pressure determine the velocity of the condensate and the initial rate of formation of the droplets. The stagnation temperature is the controlling parameter for the condensate velocity. If the stagnation temperature is too low, the condensate will start before the throat of the nozzle unless the pressure is very low. A higher plenum temperature would increase the velocity of the condensate, but too high a temperature would reduce the rate of formation unless balanced by an increase in plenum pressure.

The stagnation pressure has a very strong effect on the rate of droplet formation. If the pressure is too high, the condensate will take place too rapidly for efficient heat removal; if the pressure is too low, the condensate rate will be low and the nozzle will have to be very long to obtain a sufficient condensation fraction.

A preliminary estimate of the optimum choice of temperature and pressure for the nozzle design can be obtained from a study of Figures 7.6 and 7.7. Once the length of the nozzle required for the heat removal of the desired condensation fraction is determined, the use of Figure 7.7 will show the static temperature and pressure desired for the condensation process. Once the static temperature and static pressure are determined and the desired

condensate velocity is known, the plenum condition can be determined by use of Equation (7.9) and (7.10). The flow rate,  $\dot{m}$ , desired for a design will also be known; using this flow rate the nozzle throat area can be determined by using Equation (7.11).

Two methods of removing the condensation heat were studied. These methods were the cooling of the condensate by vapor-expansion, and the cooling of the condensate by gas injection.

The vapor-expansion cooling of the condensate at constant static temperature was found to be more efficient than vapor-expansion cooling at either constant Mach number or at constant velocity. The constant static temperature process gave the highest condensation fraction and the highest velocity.

The gas-injected system for cooling the condensate would permit the nozzle to be smaller in the condensation region and to have a higher condensation fraction than would the vapor-expansion cooling. The advantages of the gas injection system must be evaluated, taking into account the weight and the reliability penalties paid for a more complicated system. It may be found that a combination of the vapor-expansion and gas-injection system might be the best design.

Future work on aluminum colloid formation by homogeneous nucleation will be directed toward the following goals:

1. Development of a more exact method of analysis in the condensation region of the nozzle. (This preliminary analysis assumed the vapor to be a perfect gas in the condensation regions.) The analysis of the nozzle condensation region design will be made by assuming the nozzle to be the summation of many small intervals. The analysis will then be made of the conditions existing in each interval and will optimize the design for each interval.
2. A detailed design will be made of a prototype suitable for testing in the CSU vacuum chamber.
3. A computer code is being developed to aid in tasks 1 and 2 above.

## References

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- 7.2. Shapiro, H. A.: The Dynamics and Thermodynamics of Compressible Fluid Flow, Volume 1, Ronald Press Company, 1953.
- 7.3. Shin, Y. W.: "Theoretical Analysis of Condensation of Pure Vapors in Supersonic Nozzles". Masters Thesis, Colorado State University, 1965.

TABLE 7.1

Comparison of Vapor Expansion System with the  
Gas Enjection System of Cooling Condensate

ITEM	VAPOR EXPANSION	GAS INJECTION
Fraction of Vapor Condensation possible	probably less than 50% due to high weight and size penalty	100%
Additional equipment for operation of nozzle	None	Injection gas, gas storage tanks, pumps, heat exchanger, gas recovery mechanism

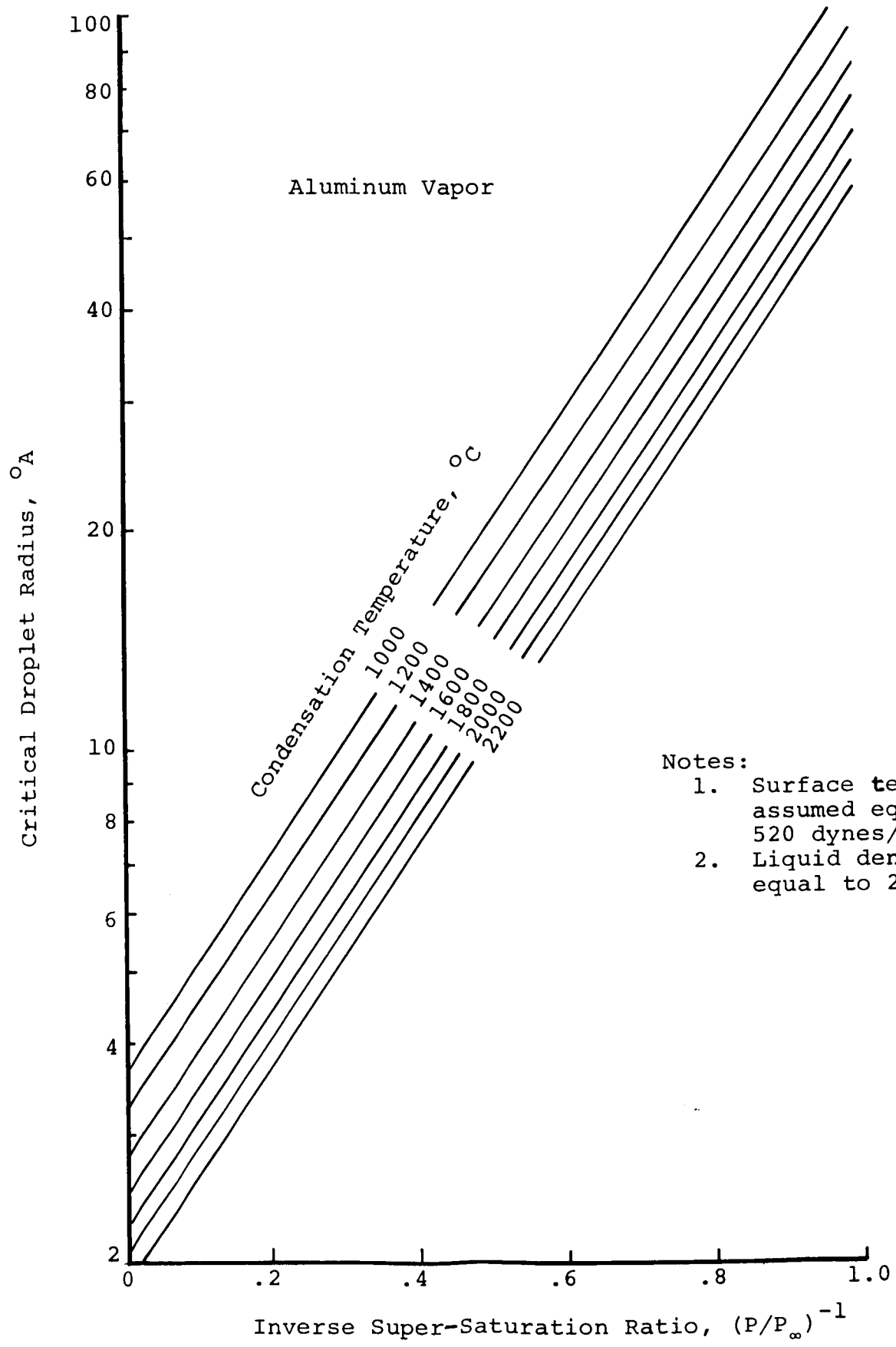


Figure 7.3. Critical Droplet Radius Versus Supersaturation Pressure and Temperature



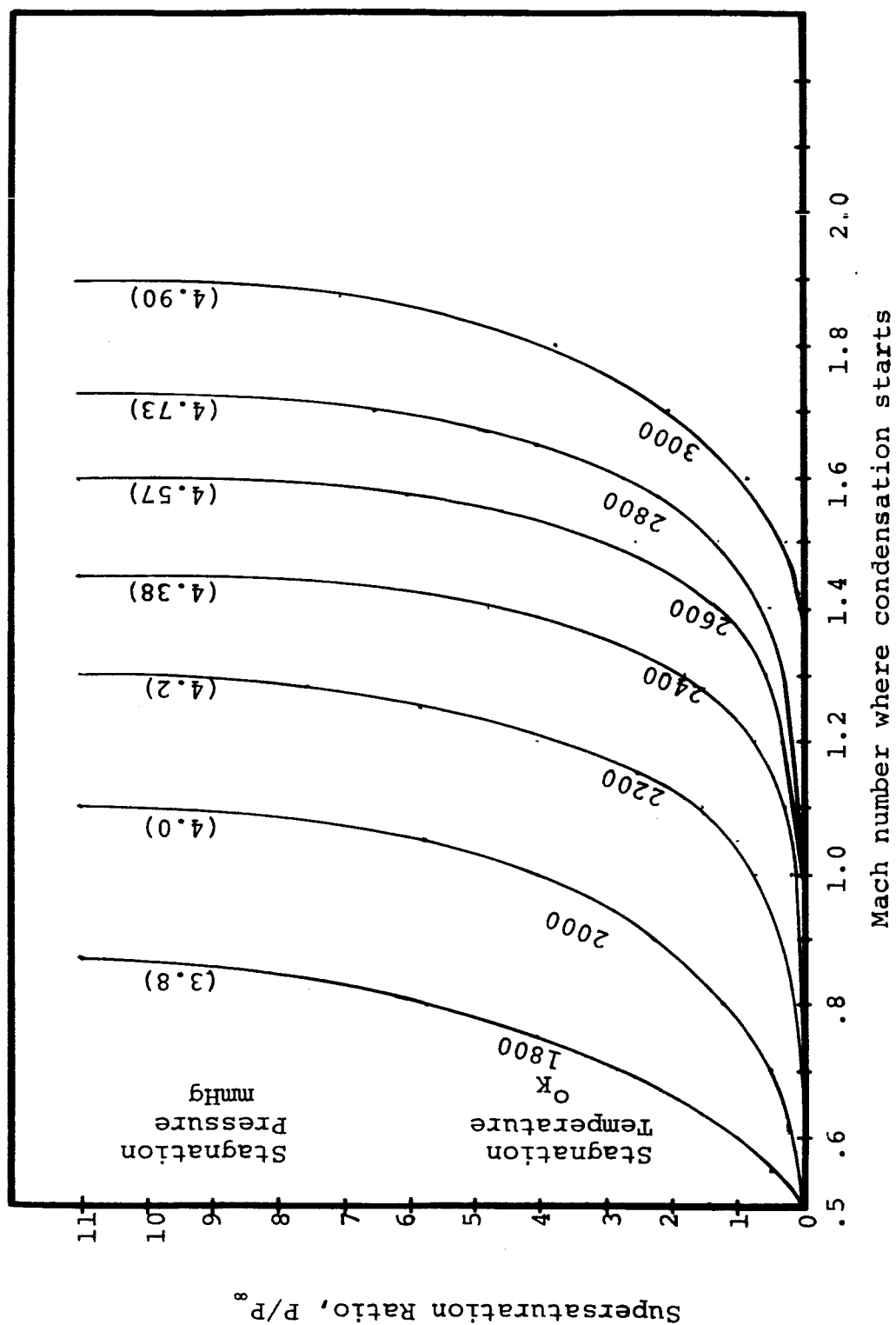


Figure 7.4. Mach Number Where Condensation Starts as a Function of Supersaturation Ratio, Stagnation Temperature and Stagnation Pressure.

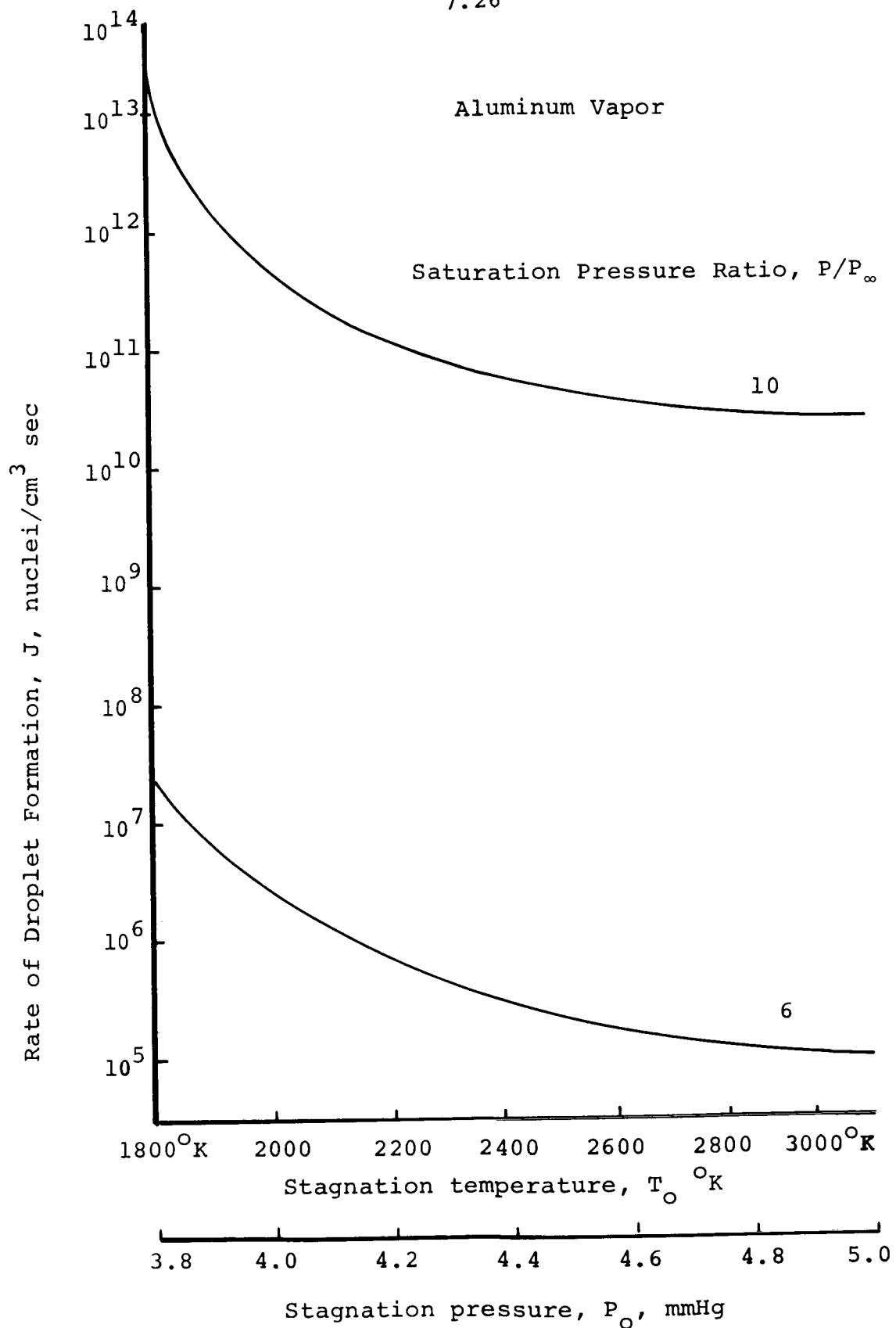


Figure 7.5. Rate of Droplet Formation as Function of Stagnation Temperature and Pressure as a Function of Saturation Pressure Ratios.

Numbers on the curves is static temperature, °C.

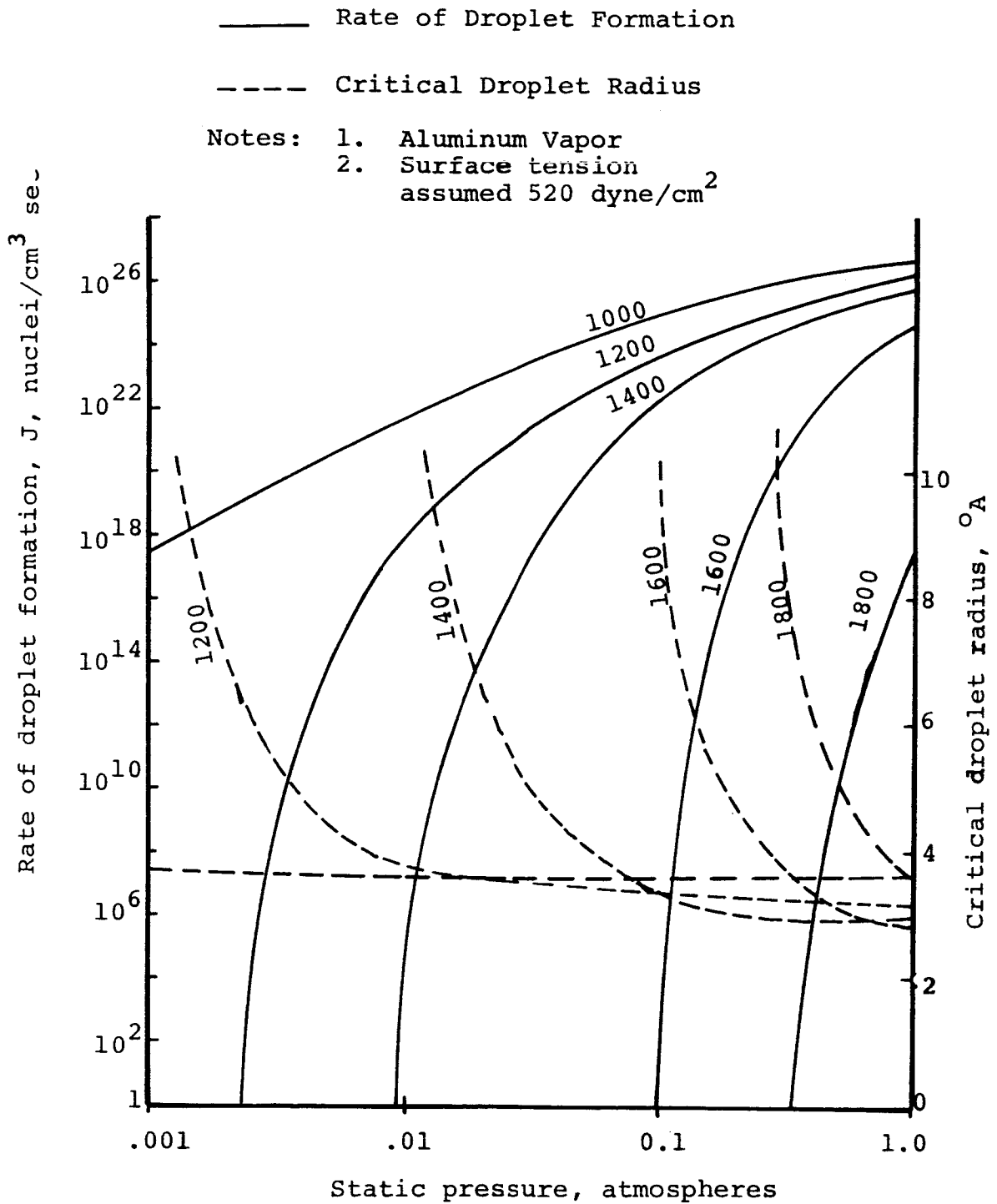


Figure 7.6. Rate of Nucleation and Droplet Radius As A Function of Pressure and Temperature

## Notes:

1. Condensation rate used shown of Figure 7.6
2. Velocity of condensate was assumed to be 1000 m/sec

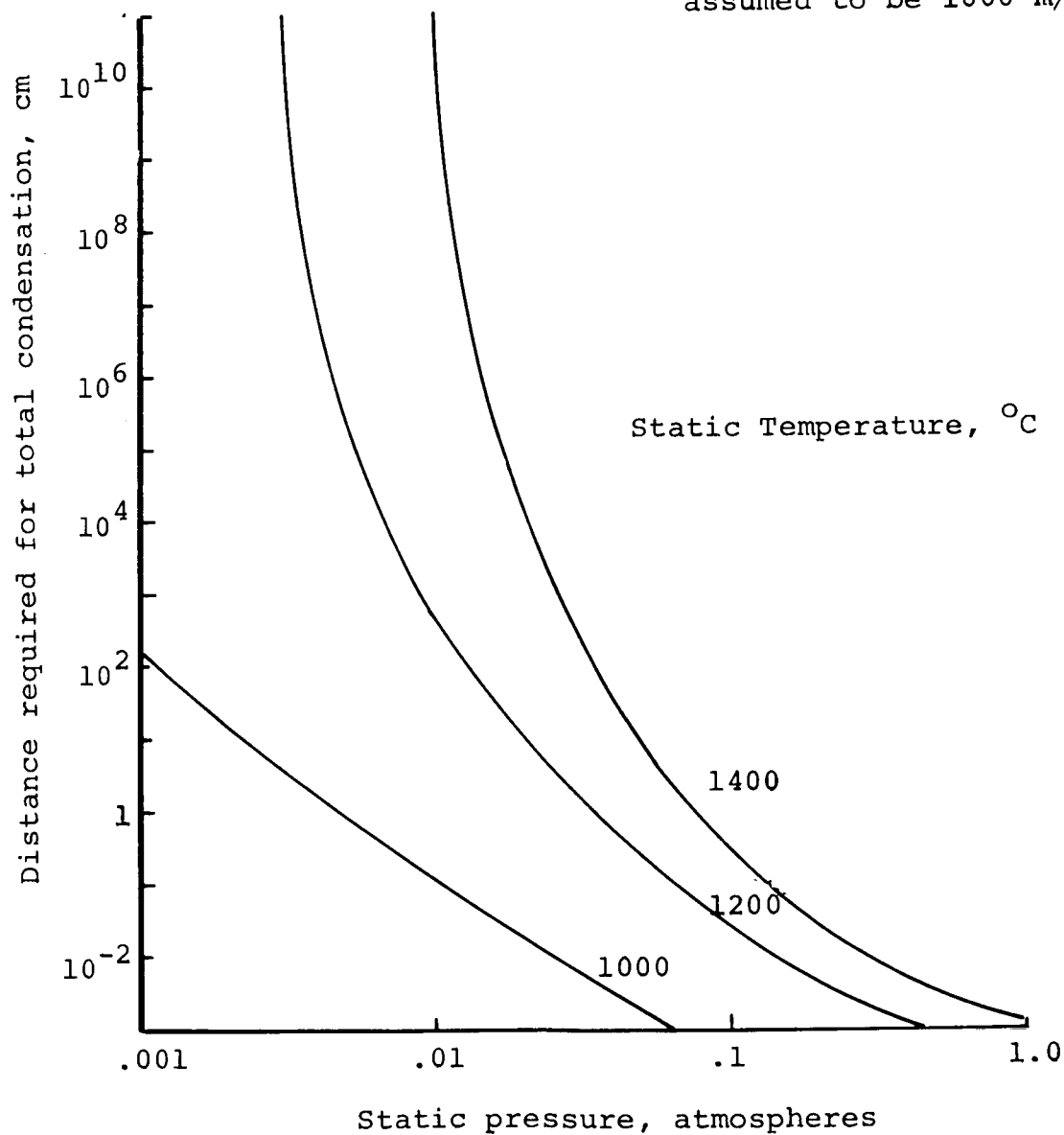


Figure 7.7. Distance Required for Total Condensation Aluminum Vapor Versus Pressure and Temperature.

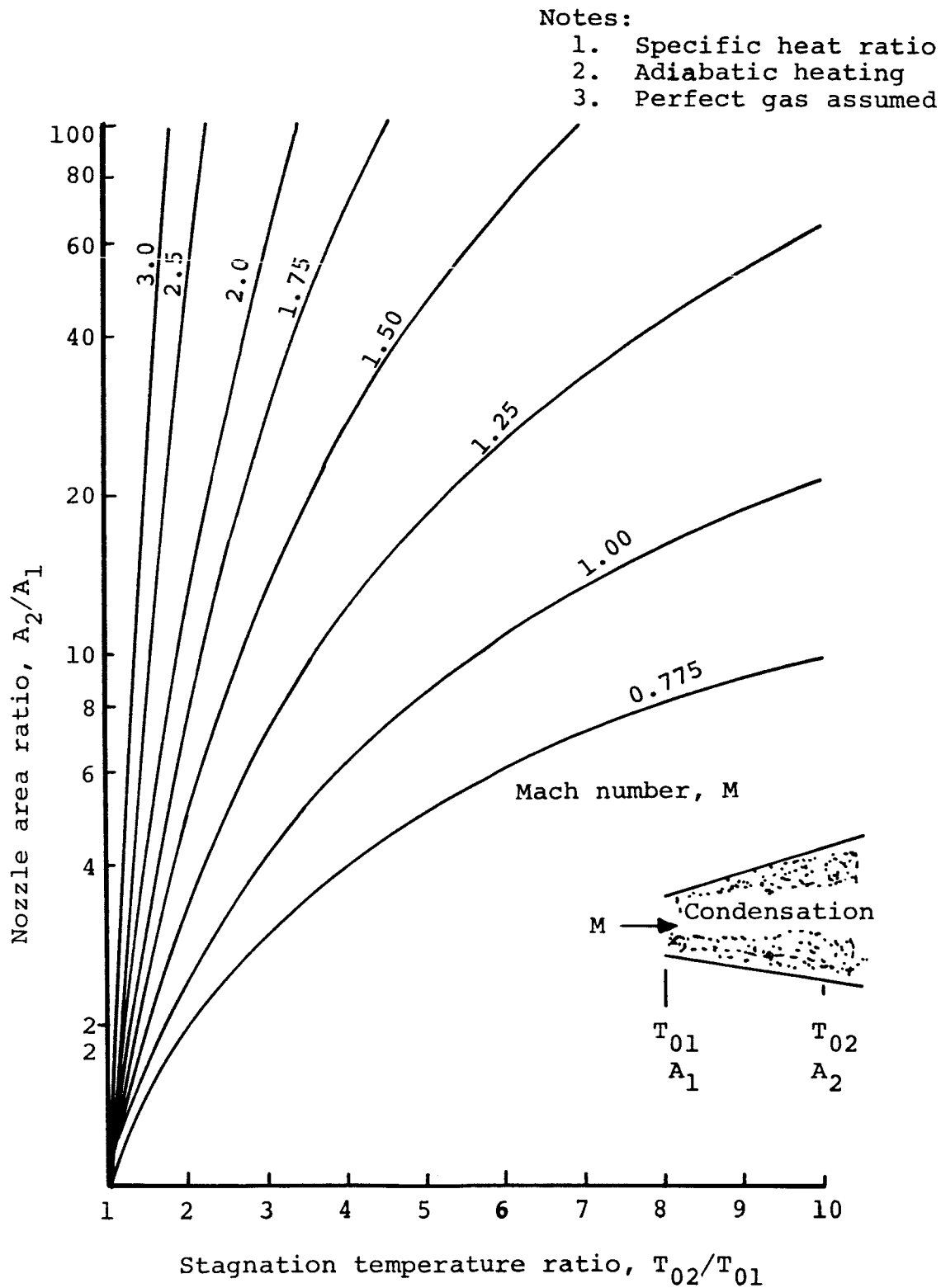


Figure 7.8. Nozzle Area Ratio Required to Hold Mach Number Constant During Condensation Heating as a Function of Stagnation Temperature Ratio.

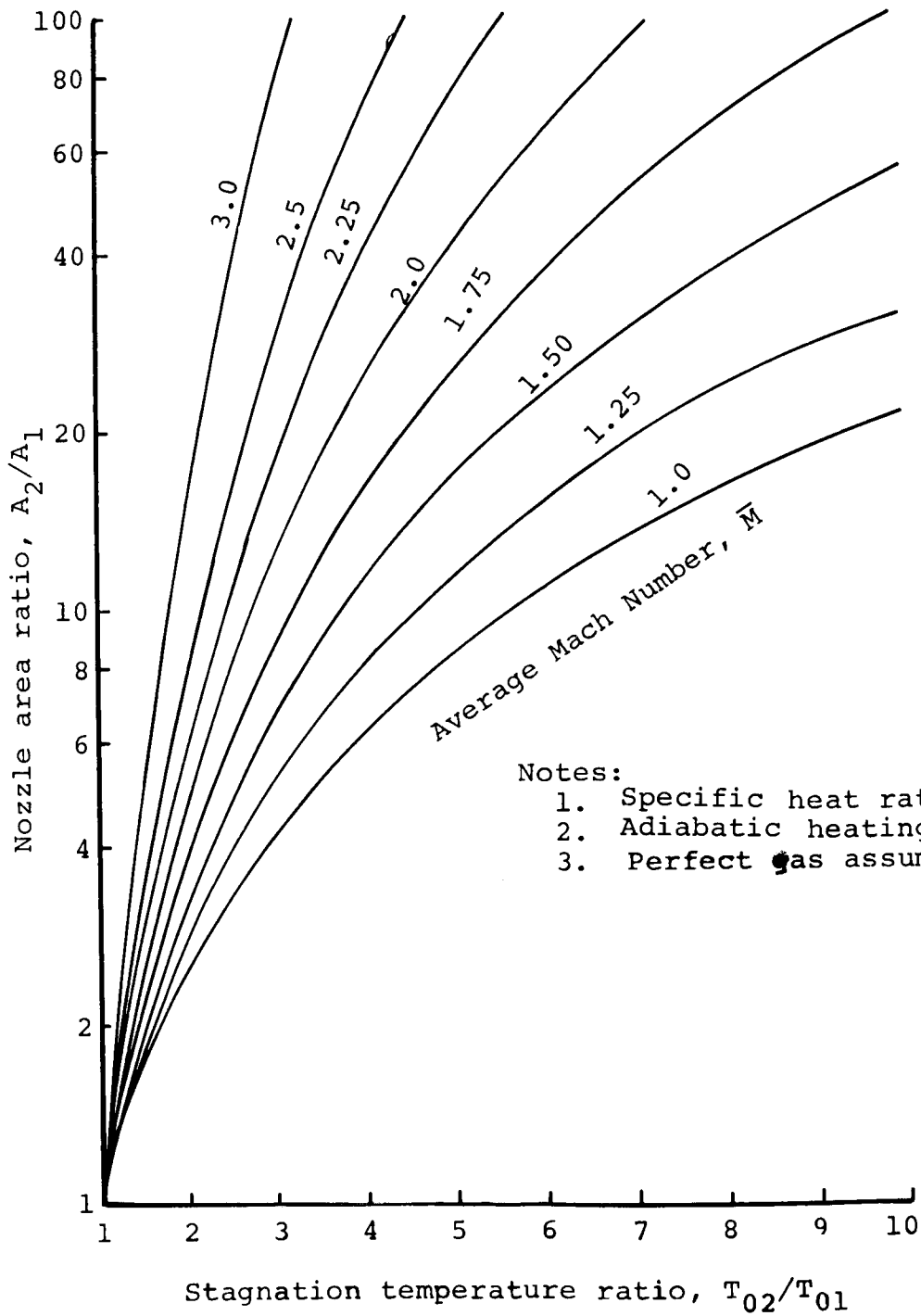


Figure 7.9. Nozzle Area Ratio Required to Hold Velocity Constant During Condensation Heating as a Function of Stagnation Temperature Ratio.

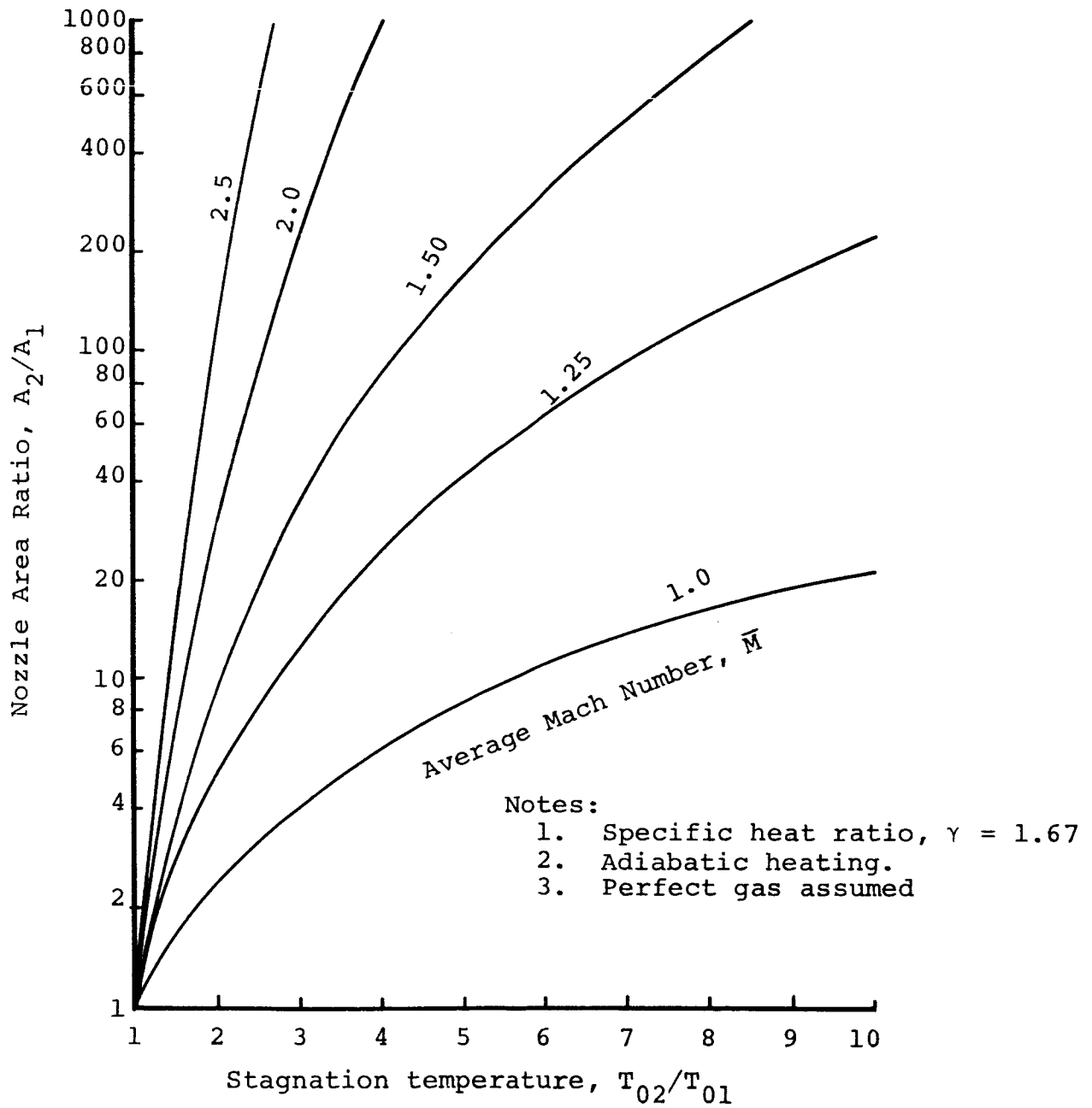


Figure 7.10. Nozzle Area Ratio Required for  $T_o$  Hold Static Temperature Constant During Condensation Heating as a Function of Stagnation Temperature Ratio